

01

NO. 02.96

97-5-56

GOVERNMENT OF JAPAN

Patent Office

KOKAI PATENT GAZETTE

(7)

(11) Early Disclosure No.: ~~Sho 52-109580~~

(43) Early Disclosure Date: September 13, 1977

(51) Int. Cl. ²	I.D. No.	(52) Japan Cl.	Intra-Office No.
B 32 B 27/28		25(9) A 11	2102-37

Examination Requested: No Number of Claims: 1 (Total of 5 pages)

(54) Title of Invention: Polypropylene-based Biaxially-oriented Composite Film with Excellent
Low-temperature Heat Sealing Properties

(21) Application No.: Sho 51-26362

(22) Application Date: March 10, 1976

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[Translator's note: The official corrections on page 465 have been incorporated in the translation.]

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1409-02-98

SPECIFICATION

1. Title of Invention

Polypropylene-based Biaxially-oriented Composite Film with Excellent Low-temperature Heat Sealing Properties

2. Claim

Polypropylene-based biaxially-oriented composite film with excellent low-temperature heat sealing properties characterized in that at least one side of a polypropylene-based stretched film is an easily heat-sealable layer composed of a mixed resin of 1-40 wt% of low-crystalline ethylene- α -olefin copolymer and 60-99 wt% of ethylene-propylene random copolymer.

3. Detailed Description of the Invention

The present invention relates to polypropylene-based biaxially-oriented composite film with excellent low-temperature heat sealing properties.

In general, biaxially-oriented polypropylene film excels in chemical and physical properties and is particularly outstanding as a transparent packaging material. But the drawback is that it shrinks when heat sealed. To overcome this drawback, a number of proposals have been made up to the present, including part coating the polypropylene film base layer, coating with an easily heat-sealable resin, laminating with an adhesive, and melt-extruding and laminating an easily heat-sealable resin. Particularly noteworthy among these proposals was a method for obtaining an easily heat-sealable biaxially-oriented composite film by laminating an easily heat-sealable resin of lower melting point than the polypropylene used for the base layer, such as ethylene-propylene random copolymer, and then strongly bonding the two by hot stretching. The heat sealability of this composite film is good, and this process is the most popular at present. However, this composite film has the following drawbacks.

(1) The melting point of the ethylene-propylene random copolymer is close to that of the melting point of the polypropylene resin used for the base layer, so a relatively high heat sealing temperature, that is, a heat sealing temperature above the melting point of this random

copolymer, is necessary. Therefore, temperature [sic] shrinkage of the stretched polypropylene base layer is stimulated during heat sealing. As a result, there is concern that wrinkles will be produced in the bags, etc., obtained, which markedly impair their attractive appearance and cause loss of commodity value.

(2) The temperature range of heat sealing is narrow, so strict temperature control is necessary when heat sealing with an automatic packaging machine or automatic bag-making machine, and sometimes the product is affected by external temperature and becomes defective.

This invention is a polypropylene-based biaxially-oriented composite film with excellent low-temperature heat sealing properties which was developed to improve the above shortcomings. It is characterized in that at least one side of the stretched polypropylene-based film that constitutes the base layer is an easily heat-sealable layer composed of a mixed resin of 1-40 wt% of low-crystalline ethylene- α -olefin copolymer and 60-99 wt% of ethylene-propylene random copolymer.

As an example of the raw material resin of the polypropylene-based resin of this invention, polypropylene resin can be mentioned, and isotactic crystalline polypropylene can be mentioned as preferred. But, of course, propylene copolymers having a melting point of at least 135°C can also be used. And as examples of the low-crystalline ethylene- α -olefin copolymer (abbreviated EO), those available on the market under the commodity names of Tafmer A4085 and Tafmer A1575, etc., manufactured by Mitsui Petrochemical Industries, can be mentioned.

The low-crystalline ethylene- α -olefin copolymers available commercially under the commodity name of Tafmer A cited as examples here are ethylene-1-butene random copolymers having a degree of crystallinity of 3-20%, ethylene content of 85-95 mol%, density of 0.86-0.91, MI (190°C) of 0.1-40, and melting point of 60-90°C.

In this instance, degree of crystallinity indicates the value obtained by dividing the peak area of the crystal peak at the diffraction location of the [100] plane according to X-ray diffraction by the intensity of the whole. Melting point is the value determined by the peak location in DSC measurement.



In addition, the ethylene-propylene random copolymer (abbreviated EP) is a well-known film-forming copolymer with an ethylene content of $\leq 10\%$ and a melting point of $\geq 135^{\circ}\text{C}$.

In this instance, the mixing ratio of the low-crystalline ethylene- α -olefin copolymer that is mixed with the ethylene-propylene random copolymer is 1-40 wt%, preferably 15-25 wt%. If less than 1 wt%, the desired results will not be provided, so this is not desirable. On the other hand, if the mixing ratio is over 40 wt%, the clarity of the stretched polypropylene film of the base layer, which was originally good, will be markedly impaired, so this is not desirable. However, practice [outside this range?] is, of course, possible depending on the application.

The mixing ratio of ethylene-propylene random copolymer is 60-99 wt%, preferably 75-85 wt%.

As examples of the method for producing the composite film of this invention, the following can be mentioned:

- (1) the method of laminating both the polypropylene based-resin and the blend of ethylene-propylene random copolymer and low-crystalline ethylene- α -olefin copolymer inside the extruder die or near the outlet thereof, or in the molten state, and then biaxially orienting consecutively or simultaneously;
- (2) the method of forming both into solid films, superposing these, and laminating, or melt-extruding and laminating one on a solid film of the other and then biaxially orienting consecutively or simultaneously; and
- (3) the method of stretching the unstretched polypropylene-based film uniaxially in advance, and then melt-extruding and laminating the mixture of ethylene-propylene random copolymer and low-crystalline ethylene- α -olefin copolymer on that film, or molding that mixture into a film, stacking, laminating, and then stretching in the direction perpendicular to the above-mentioned direction.

Lamination can also be carried out by the hot melt process, etc., instead of the aforementioned melt-extrusion lamination.

As the ideal method among those mentioned above, the method of first stretching the unstretched polypropylene-based film by ≥ 1.2 times in the machine direction with metal rolls,

etc., next melt-extruding and laminating the mixture of ethylene-propylene random copolymer and the low-crystalline ethylene- α -olefin copolymer, and then stretching by ≥ 1.3 times in the transverse direction with a tentering frame can be mentioned.

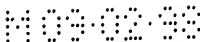
The stretching temperature is preferably above the melting point of the low-crystalline ethylene- α -olefin copolymer when it exhibits the lower melting point of the two resins of the easily heat-sealable layer, and the easily heat-sealable layer is preferably in an essentially non-oriented state. By this, the desired effects are further enhanced. If the easily heat-sealable layer is in an oriented state, it is conceivable that it will also shrink during heat sealing, so this can hardly be called desirable. However, this practice is possible depending on the application.

If the film surface of the easily heat-sealable layer of this invention poses a problem in terms of sliding resistance, etc., that can be solved by increasing the loading of the ethylene-propylene random copolymer, but besides that, polyethylene, polypropylene, or other suitable resins compatible with the low-crystalline ethylene- α -olefin copolymer may be added. Also, if necessary, fillers and lubricants may be added. In this case, silica-based fillers and oleamide-based lubricants are especially ideal. In addition, well-known antistatic agents, antifoggants, and the like may also be added as necessary.

Providing the easily heat-sealable layer of this invention on one side of the base layer will suffice, but, of course, it can also be provided on both sides of the base layer, depending on the application.

The properties of the composite film of this invention will now be explained using the appended drawings.

Figure 1 shows the relation between heat seal strength and heat sealing temperature for various mixing ratios of the ethylene-propylene random copolymer and low-crystalline ethylene- α -olefin copolymer of the easily heat-sealable layer. As is clear from this figure, as the mixing ratio of the low crystalline ethylene- α -olefin copolymer of this invention increases, heat seal strength rises, and in addition, heat sealing properties are good even at low temperatures such as 110°C. As a result, wrinkles and cracks are not generated by heat shrinkage of the stretched



polypropylene layer during heat sealing, and packaging that is attractive and strong and improves the commodity value of the packaged product becomes possible.

The heat sealing temperature range extends over the broad range of 110-140°C, so when used during automatic packaging and in automatic bag-making machines, strict control of the temperature is not necessary, and control is extremely easy.

By contrast, the temperature at which heat sealing begins in the case of the conventional product, where the easily heat-sealable layer is ethylene-propylene random copolymer alone, is high, as is clear from the figure, so it lacks low-temperature heat sealability. Also, the temperature range in which heat sealing is possible is very narrow, and for this reason, the conventional product has the various drawbacks indicated above.

The method of determining heat seal strength was to overlay the easily heat-sealable surfaces of the composite film, apply heat and pressure with a bar-type heat sealing machine at a pressure of 1 kg/cm² for 1 sec, and then subject the 1-cm wide specimen obtained to a peel test at a peeling rate of 200 mm/min and peeling angle of 180°. Heat seal strength is shown in units of g/cm, as below.

Figure 2 shows the clarity at various mixing ratios of the two resins used in the easily heat-sealable layer and is indicated by turbidity (%). Good clarity means the same thing as low turbidity. As is clear from this figure, clarity worsens sharply when more than 40 wt% of low-crystalline ethylene-propylene copolymer is used, and the material can hardly be used in applications as a transparent packaging film.

The invention will now be explained with practical examples.

Practical Example 1

Isotactic polypropylene resin was extruded in sheet form with a T-die extruder and passed through heated rolls, thereby stretching it in the machine direction to an effective stretching ratio of 5 times. On one side of this uniaxially oriented sheet was melt-extruded and laminated a mixed resin of 80 wt% ethylene-propylene random copolymer and 20 wt% low-crystalline ethylene- α -olefin copolymer, which was an ethylene-1-butene random copolymer with a degree of crystallinity of 3-20%, ethylene content of 85-95 mol%, MI (190°C) of 0.1-40, density of



0.86-91, and melting point of 60-90°C, available commercially under the name Tafmer A4085 from Mitsui Petrochemical Industries. This composite sheet was stretched to an effective ratio of 9 times in the transverse direction by passing it continuously through a tentering frame to obtain biaxially-oriented composite film. In this instance, the thickness of the propylene layer was approximately 35 μm , and the thickness of the easily heat-sealable layer composed of a mixed resin of ethylene-propylene random copolymer and low-crystalline ethylene- α -olefin copolymer was approximately 5 μm .

Practical Examples 2-5

Biaxially-oriented composite films were obtained by the same method as in Practical Example 1 except for changing the mixing ratios of the ethylene-propylene random copolymer and low-crystalline ethylene- α -olefin copolymer used as the easily heat-sealable layer as shown in Table 1.

Table 1

	PE 2	PE 3	PE 4	PE 5
% EP	95	90	70	60
% EO	5	10	30	40

PE = Practical Example

Comparative Example 1

Biaxially-oriented composite film was obtained by the same method as in Practical Example 1 except for using only ethylene-propylene random copolymer as the easily heat-sealable layer. The relation between the heat sealing temperature, heat seal strength, and turbidity of these is shown in Table 2.



Table 2

			PE 2	PE 3	PE 1	PE 4	PE 5	CE 1
Composition of easily heat sealed layer EO:EP			5:95	10:90	20:80	30:70	40:60	/100
Heat seal strength (g/cm)	Heat sealing temp. (°C)	110	50	150	310	350	450	0
		115	280	350	380	430	500	0
		120	430	480	510	560	580	350
		125	480	520	550	650	690	360
		130	500	560	600	700	750	380
		140	550	600	650	720	800	400
Turbidity	(%)		1.6	1.8	2.1	3.0	5.5	1.6

PE = Practical Example CE = Comparative Example

As is clear from Table 2, it was found that the composite films of the practical examples were excellent films that could be heat sealed at a low temperature, and therefore the heat sealing temperature range was also very broad and heat seal strength was high. As opposed to these, the heat-sealable temperature of the film of Comparative Example 1, a conventional product, was high, and the usable heat sealing temperature range was very narrow. With respect to turbidity, the film of Practical Example 5 showed a rather poor value, so when the mixing ratio of low-crystalline ethylene- α -olefin copolymer was over 40 wt%, the film was unsuitable for use as transparent packaging film.

4. Brief Explanation of the Figures

Figure 1 is a graph showing the relation between the heat seal strength and the heat sealing temperature of the composite films. Figure 2 is a graph that shows the relation between the mixing ratio of the resins used and turbidity.

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Figure 1

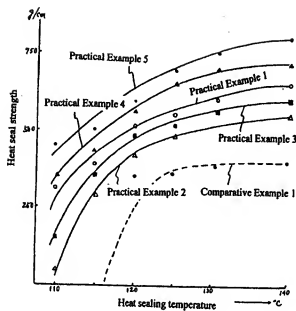


Figure 2

